Formation of Mixed-layer Sulfidehydroxide Minerals from the Tochilinite-Valleriite Group During Experimental Serpentinization of Olivine

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Minerals in the tochilinite-valleriite group (TVG) are composed of a mixture of sulfide and hydroxide layers and have been reported as accessory components in many serpentinites. Despite their low abundance, TVG minerals have the potential to provide valuable insights into local environmental conditions during serpentinization, provided that the circumstances under which they form can be accurately constrained. Heretofore, however, TVG minerals had never been reported in laboratory simulations of serpentinization, leaving the conditions under which they form largely uncertain. We will report the formation of members of the TVG mineral group (specifically, Cu-rich valleriite and Ni-rich haapalaite) during laboratory serpentinization experiments conducted at 300°C and 328°C. The experiments were conducted by reacting olivine \pm a small amount of FeS (500 ppm) with artificial seawater for 166 days. Isotopically labeled sulfate (~20 mol% ³⁴SO₄) was used in order to trace the fate of S during the experiments. Reacted fluids became mildly acidic, and contained low levels of Mg and Si as well as 1-3 mmolal SO₄. Elevated levels of dissolved hydrogen (H_2) were generated during the experiments; however, the amounts of H₂ produced were significantly lower than observed in similar experiments conducted without SO4 present, reflecting substantial reduction of sulfate. About 14% of the olivine reacted during both experiments, producing primarily chrysotile, brucite, and magnetite. At 300°C, the reacted solids included trace amounts of haapalaite along with pentlandite, while at 328°C valleriite and an unidentified Ni,Fe-sulfide were observed. Sulfur isotope analysis indicates that both the TVG minerals and sulfides incorporated a significant amount of S from sulfate reduction. Overall, the results suggest that some TVG minerals in serpentinites could have formed at highly elevated temperatures under reducing, mildly acidic-to-circumneutral conditions, and may incorporate some S from reduction of seawater sulfate.